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Observations of nucleation mode particles formation and growth on Mount Huang, China

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Abstract

The growth of nucleation mode particles is analyzed using the measured data of aerosol particle size distribution, trace gas and meteorological parameters collected on Mount Huang, China from September 22 to October 28, 2012. The new particle formation (NPF) events were observed on average 18.2% of all measurement days and all occurred before the noon of sunny days. Compared to non-event days, the wind speed, SO_2 and O_3 concentrations were higher on NPF days, whereas the temperature and relative humidity (RH) were at comparatively lower levels. The concentrations of small particles (10~20nm) increased first, and then the larger particles (20~50nm) concentrations increased over time. The peaks of large aerosol particle concentration were lower than that of small ones and the average growth rate (GR) of the newly formed aerosol particles was 3.58nm h^{-1}. SO_2 concentration peaked before nucleation mode particle number concentration reached to its maximum. SO_2 participated in the particles nucleation as a kind of precursor and influenced the growth of newly formed particles. In our studies SO_2 concentrations were found to be the main influencing factor of the aerosol particle growth rate, so the aerosol particle growth rate was higher with higher concentration of SO_2. The basis for observation and parameterization schemes in this work will contribute to the database enrichment and model simulations of nucleation mode particle growth over this region.

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Keywords: New particle formation; growth rate; trace gas; Mount Huang

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1. Introduction

Ubiquitous in the atmosphere, atmospheric aerosol particles play an important role in the earth's radiative balance through the absorption and scattering of the incoming radiation. The aerosol particles exert their influence in several ways, partially through the indirect climate effect by acting as cloud condensation nuclei (CCN) or ice nuclei (IN) changing the characteristics and lifetime of clouds. Meanwhile the size distribution and concentration of the aerosol particles, together with their composition and so on, affect the visibility and human health through their inhalation.

New particle formation is an important source of atmospheric aerosols, and is a key factor influencing the properties of aerosol particles. New particles are formed by nucleation of non-volatile or low-volatile gas-phase compounds, emitted from either biogenic or anthropogenic sources, followed by growth into small particles [1]. There are five nucleation mechanisms [2], binary nucleation of H_2SO_4 -H_2O [3], ternary nucleation of H_2SO_4 -H_2O involving ammonia amines [4], nucleation of H_2SO_4-H_2O assisted by organic acids [5], nucleation of iodine oxides [1] and ion-induced nucleation [6].

In recent years the importance of NPF has motivated a series of world-wide observations in different stations, which span from clean continent [7-9], polluted continent [10-11], marine [12] and boundary layer [13-15]. The observations in China were focused on Beijing [16-19], Yangtze River delta [20-21], Pearl River Delta [22] and Hong Kong [23-24]. Kulmala et al [25] summarized the results of previous observation studies, they concluded that the formation rate of 3-nm particles often spans in the range of 0.01~10cm^{-3} s^{-1} in the boundary layer. However, in urban areas formation rates are often higher (up to 100$cm^{-3}s^{-1}$), and rates as high as 10^4~10^5cm^{-3} s^{-1} have been observed in coastal areas and industrial plumes. Typical particle growth rates vary in the range of 1~20nm h^{-1} in mid-latitudes due to the diversity of the temperature and the availability of condensable vapor. Over polar areas the growth rate can be as low as 0.1nm h^{-1}. The growth rates in Beijing [16-19], Yangtze River delta [20-21], Pearl River Delta [22] and Hong Kong [24] are respectively in the range of 0.1~11.2nm h^{-1}, 4.8~7nm h^{-1}, 2.2~19.8nm h^{-1} and 1.5~8.4nm h^{-1}. The formation rates in Beijing [16-19] and Pearl River Delta [22] are 1.1~81.4cm^{-3} s^{-1} and 0.5~5.2cm^{-3} s^{-1}. In general, one conclusion can be drawn that the growth rates in urban areas are higher than that in clean areas.

More observations in different types of environments in China are still needed to supplement the research of new particle formation. In the present paper we analyze the characteristics of new particle formation on Mount Huang, a background site located in eastern China. Previous studies [26-27] show that its background aerosol number concentrations are low and this site can be classified as clean continental area. The concentration of pollutant is mainly influenced by mountain-valley breeze [26], which is greater than the impact of human activity and atmospheric structure. Due to the low concentration of the background atmospheric aerosols the gaseous pollutants can stay in the atmosphere for longer time growing to the observable size range after nucleation and won't be removed by collision elimination. The results obtained in this study will enrich the database of new particle formation observation at background site in eastern China and provide the basis for observation basis and parameterization schemes for new particle formation simulations by numerical models.

Nomenclature

CCN	cloud condensation nuclei
GMD	geometric mean diameter
GR	average growth rate
IN	ice nuclei
NPF	new particle formation
RH	relative humidity
WPS	Wide-Range Particle Spectrometer
WS	wind speed

2. Methods

2.1. The sampling site, instruments and data analysis

Mount Huang is situated in the south of Anhui province in China. Observations were conducted on Yungu Temple on Mt. Huang at 869m a.s.l. (30.12°N, 118.18°E) , which is surrounded by rich vegetation and we can eliminate the impacts of anthropogenic pollution sources. The sampling time is from September to October with low background particle number concentration favoring to study the characteristic of new particle formation and growth.

Number concentrations and size distributions measurements of ambient aerosol particles were continuously performed by Wide-Range Particle Spectrometer (WPS) in a range of diameters from 10nm to 10μm, during 22 September to 28 October in 2013, 37 days in total. WPS is an instrument that can measured particles in 67 bins, with a time resolution of 5 minutes. The dryer made the relative humidity of the air going to the sampler below 40% to eliminate contamination of the working fluid. The meteorological parameters such as the atmospheric temperature, pressure, humidity, wind and the rainfall were also continuously measured by the automatic meteorological station with a time resolution of 1 minute. In order to explore the effect of the trace gas to the new particle formation, the concentrations of the SO_2 and the O_3 were measured at the same time. The data in 30mins after the WPS started were rejected in order to ensure the data quality. And we removed the data collected during the instrument malfunction and the measurements which were observed in less than continuous 12 hours a day. Similarly meteorological data and trace gas data were also quality control data, excluding the data with large error due to machine switch and equipment failure. We get 8806 samples and 33 effective observation days in total.

2.2. New particle formation (NPF) events classification

We use the criteria defined by Dal Maso et al [9] through the study of an 8-year- continuously observation at a boreal forest measurement site at Finland. For a day to be classified as an NPF event, the criteria are as follows:

1. A distinctly new mode of particles must appear in the size distribution;
2. The mode must start in the nucleation mode size range;
3. The mode must prevail over a time span of hours;
4. The new mode must show signs of growth.

According to this classification we can identify which days can be called new particle formation (NPF) days. Our dataset comprised 33 days in total of which the event days amounted to 6 days, including 30 September, 11 October, 12 October, 17 October, 18 October and 23 October. 3 days of the total, which don't fulfill the criteria of events or non-events, are undefined days and the rest of the days (24 days) are non-event days, with low particle number concentration and no NPF events.

2.3. The calculation of the particle growth rate

The particle growth rates between two sizes classed can determine the speed of the particles switching from small diameters to large sizes during the whole process of particle growing. The new particle growth rate is computed using the formula ($GR = \Delta D_m / \Delta t$), where $\triangle D_m$ is the increased diameter and $\triangle t$ is the lasting time [28].$GR_{10\sim20}$, $GR_{20\sim30}$ $GR_{30\sim40}$ and $GR_{40\sim50}$ represent respectively the growth rate of aerosols whose diameters range in 10~20nm, 20~30nm, 30~40nm and 40~50nm separately.

3. Results and discussion

3.1. Statistics of new particle formation (NPF) event

A total of 6 NPF events were found during the observing period (Table.1). The start time is specified as the time when the aerosol number concentrations in the diameter of 10~15nm ($N_{10\sim15}$) bumped up and the increment of $N_{10\sim15}$ is 10^2 cm^{-3}. The end time is specified as the time when the particle diameter would no longer increase. The earliest

start time was 8:45 and the latest was 10:53, both were at the time when the solar radiation began to increase. Meanwhile the earliest end time was 17:20 and the latest end time was 19:47. That was the boundary between afternoon and night time, and the solar radiation has been very weak, which indicated that the new particle formation- growth process required strong solar radiation [29]. The averaged durations were 8.8 hours with a minimum of seven hours and a maximum of ten hours We could classify all of the 6 NPF events as the class I defined by Hamed et al [11]. Class I events show intensive and clear formation of small particles with continuous growth to large particle sizes that lasts from seven to ten hours with an average of about eight hours, this indicated the NPF events on Mount Huang had the unifying characteristics. Otherwise each NPF events were not the same, the particle growth rates ranged from 2.29nm h^{-1} to 4.27nm h^{-1} with an averaged GR of 3.58nm h^{-1}. This GR was similar to the ones in clean areas [24], but lower than those of polluted areas [16-17]. The geometric mean diameter (GMD) had a good linear relationship with time in each NPF event, and the correlation coefficients were all above 0.94 with an average of 0.96 as is showed in Fig.1. We also found that the events with a higher GR ultimately corresponded to a larger particle final diameter, showing a positive correlation of the final particle diameter and GR.

Table 1. Case statistics of aerosol particle growth during observation

Date	Stat time	Duration	End time	Primary diameter(nm)	Final diameter(nm)	GR(nm h^{-1})
2012/09/30	9:10	9:49	18:59	13.79	55.70	4.27
2012/10/11	10:53	8:07	19:00	13.79	47.85	4.20
2012/10/12	10:14	9:33	19:47	11.95	44.37	3.40
2012/10/17	10:03	7:17	17:20	14.81	44.37	4.06
2012/10/18	9:55	8:22	18:17	13.79	32.94	2.29
2012/10/23	8:45	9:59	18:44	11.95	44.37	3.25

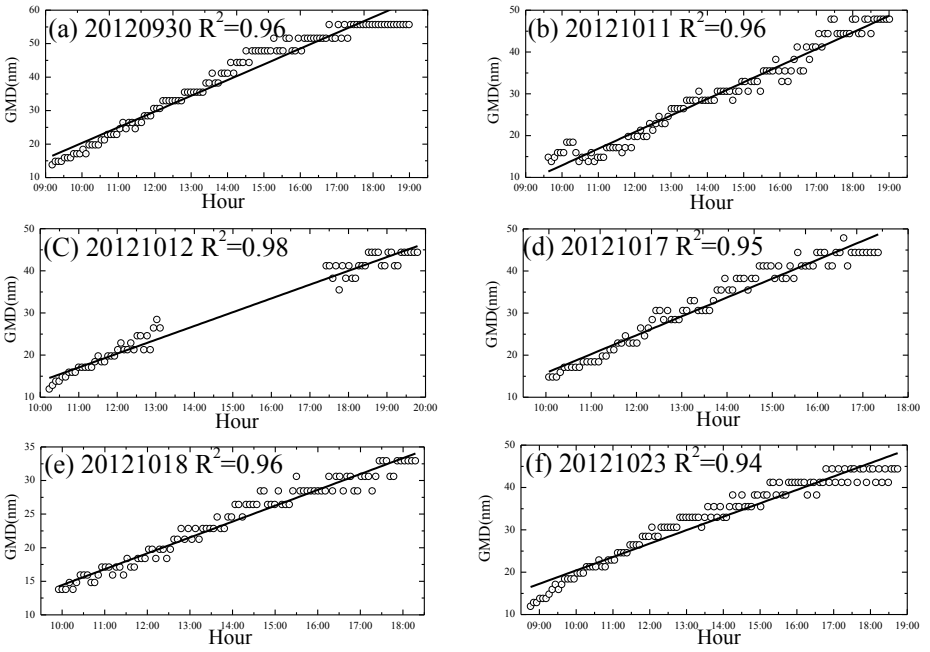

Fig1. Tendency of the geometric mean diameter during particle growth: (a) September 30；(b)October 11；(c) October 12; (d) October 17; (e) October 18; (f) October 23.

3.2. Analysis of NPF events characteristics

3.3.1. Case study of new particle formation

Figure 2 illustrates a new particle formation event on October 23. The number concentration of the aerosols smaller than 50nm was sustained at a low value (100 cm^{-3} or less) during 00:00~08:00(Fig 2b), indicating a really low ultrafine particle concentration in the atmosphere before the NPF event. The aerosol number concentration in the diameter range of 10~20nm ($N_{10\sim20}$) begun to increase at 08:45 and reached the peak after just one hour with a tenfold increase in the concentration. Figure 2a shows what is informally called the nucleation "banana". The number concentration of aerosols in a diameter ranging from 20 nm to 30nm ($N_{20\sim30}$) showed a sudden increase at 09:10 with a great increment of two orders of magnitude. The number concentration of aerosols in the diameter of 30~40nm ($N_{30\sim40}$) begun to rise at 10:00 and reached to the maximum after 3hours and maintain a sustainment at about 3000 cm^{-3} in the following 1hour. The aerosol number concentration in the diameter range of 30~40nm ($N_{30\sim40}$) showed a tiny increase at about 12:00 and reached the peak after 4hours.

The increment of aerosol number concentration shows 3 features: 1. the start time of aerosol number concentration growth delays with the increase of aerosol diameter; 2. the maximum of aerosol number concentration decreases as the aerosol diameter increases; 3. the rate of increase in aerosol number concentration decreases with the increasing aerosol diameter. The supersaturated gas phase pollutants turn to aerosol particles smaller than 10nm by condensation in the background atmosphere. More and more aerosol particles grow into larger diameters through coagulation and condense on supersaturated gas phase pollutants, that is why $N_{20\sim30}$ showed the tendency of increment. The concentration increase of aerosols larger than 20nm is based on the condensation and coagulation of small aerosols. Collectively the NPF event leads to an explosive growth in the number concentration nucleation mode aerosols, providing a basis for the quantitative increase in the number concentration of Aitken mode aerosol. Table 1, Fig 1 and 2a show that the diameters of newly formed aerosols on Mount Huang don't reach 100nm scale, which is different from urban areas [16-18], indicating that the NPF event is a local phenomenon[11]. The NPF event may be influenced by background aerosol number concentration, local emission and long-range transmission pollutant.

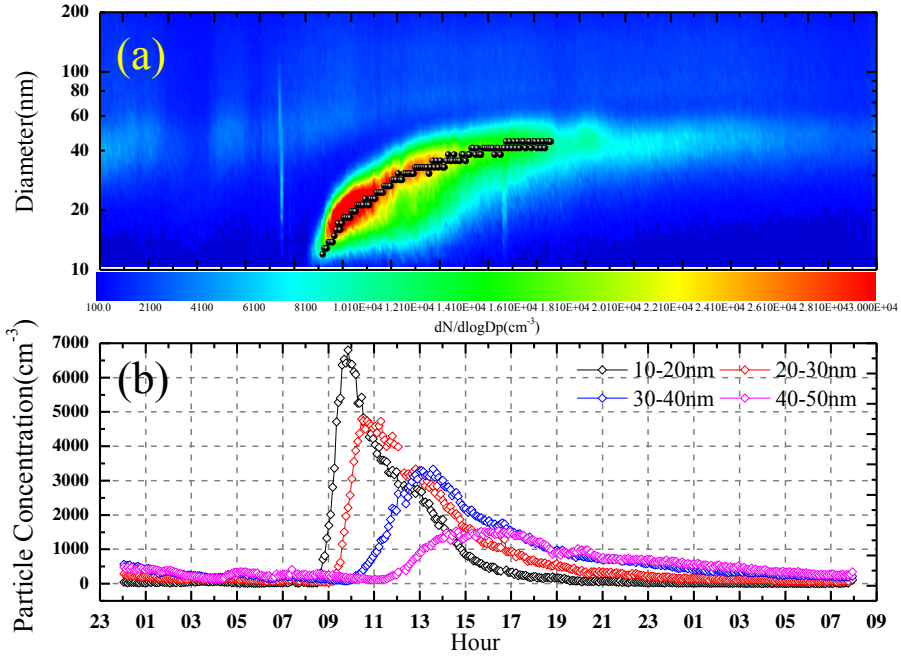

Fig 2. (a)Contour plot of new particle formation and growth on 23 October 2012(black plots present particle geometric mean diameter); (b) hourly variation of 4 different sizes aerosol number concentration

Fig 3 illustrates the hourly variations of meteorology elements and trace gases on 23 October. The wind direction appeared to influence the occurrence of particle nuclaeation and growth events. The wind direction turned from north to south at 09:00 and turned second time at 15:00, which reflect the phenomenon of mountain-valley breeze, a main breeze in the mountain. The wind speed (WS) maintained fairly low (below 1m $s^{-1)}$ the whole day, which favored the stay of pollutant at the observation station and avoided the newly formed aerosol particles from being blew away. The relative humidity (RH) sustained below 75% all day long and decreased to less than 55% after the NPF event occurred. Studies show a negative correlation between the occurrence of NPF events and RH [18, 23], because low RH contributed to the condensation of the gas phase pollutant in the atmosphere. The number concentration of nucleation mode aerosols (N_{nuc}) and Aikten mode aerosols (N_{ait}) were respectively in the magnitude of 10^2 cm^{-3} and 10^3 cm^{-3} both before and after the NPF event. A peak in N_{nuc} and N_{ait} appeared during NPF event, indicating NPF event is the main factor of N_{nuc} and N_{ait} increase.

The SO_2 concentration began to rise at 06:00 and reached the maximum at 09:00 which is 1hour earlier than the N_{nuc} reached its peak. Studies show that oxidation process of SO_2 to sulphuric acid is the main step for new particle formation and the growth of newly formed aerosol particles [30]. The observation results in this study corroborate the process. When SO_2 gas reached the oxidation condition, the chemical reaction with O_3 is as follows:

$3SO_2 + 3H_2O + O_3 = 3H_2SO_4$ (1). Weber[31] indicated that H_2SO_4 was a vapor precursor of the newly formed particles, and the MSA contributed little to new particle formation. In this study H_2SO_4 may also be the main precursor of NPF event, but it's still remained to be confirmed.

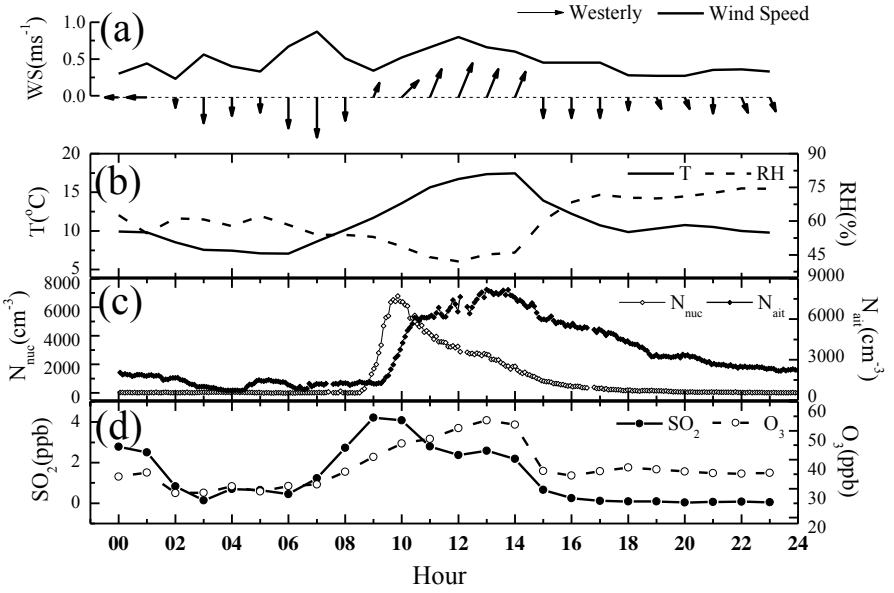

Fig 3. Hourly variations of meteorology elements and trace gases on 23 October 2012(a. wind direction and speed; b. temperature and relative humidity; c. aerosol number concentrations on nucleation mode and Aitken mode; d. concentrations of SO_2 and O_3)

3.2.2 Statistical characteristics of aerosols

NPF events contribute to the number concentration of ultrafine aerosols, while the number concentration in non-NPF event days may reveal other possible sources of aerosols. The number concentration of aerosols in the range of 10nm~10 μ m is $2.3\times10^3$$cm^{-3}$ on average, which is in the same magnitude with that on the top of Mount Huang[26~27,32]. We made a comparison of aerosol number concentrations between NPF days (event day) and non-NPF days (non-event day) (Fig 4) which has shown evident differences. The number concentrations of aerosols in nucleation mode (10~20nm), Aitken mode (20~100nm) and particles in the range of 10nm~10 μ m (N_{nuc}、N_{ait}、N_{tot}) are all higher on event days, while the aerosol number concentration in accumulation mode (N_{acc}) shows the opposite characteristic.

On event days N_{nuc} began to increase at 05:00 and showed a maximum increment with $1.6\times10^3$$cm^{-3}$ during

08:00~09:00, and then reached the peak value $2.9\times10^3 cm^{-3}$ during 10:00~11:00. On non-event days N_{nuc} was always below $1.5\times10^2 cm^{-3}$ and reached the maximum at 15:00. N_{ait} was also much higher on event days with an increment at 06:00 and a maximum increment of $1.7\times10^3 cm^{-3}\ h^{-1}$ during 09:00~10:00. The maximum was $7\times10^3\ cm^{-3}$ at 13:00, which is 2.4 times of the peak value of N_{nuc}. On non-event days N_{ait} was also sustained at a low value and reached its maximum of 1.1×10^3 at 17:00. N_{acc} showed the same trend on both conditions during 01:00~11:00, but after 12:00 the tendency was quite different. N_{acc} kept increasing after 11:00 on event days and reached maximum at 20:00, whereas it increased first and then decreased on non-event days. The accumulation mode aerosols are mainly derived from one emission, Fig 4c illustrates that before 11:00 the source on both conditions may be the same and N_{acc} on event days kept increasing due to NPF process through the coagulation of new particles smaller than 100nm in diameter.

N_{ait} accounts for the largest proportion of N_{tot} on event days while N_{acc} accounts for the smallest. On non-event days the value of N_{ait} and N_{acc} are both $8\times10^2\ cm^{-3}$. The fact that N_{acc} on non-event days is much higher than event days illustrates that NPF occurs in clean atmosphere with low aerosol number concentration. High aerosol number concentration may lead to the chemical reaction between trace gas and suspended aerosols, meanwhile the newly formed aerosol particles may disappear through the coagulation with the suspended aerosols.

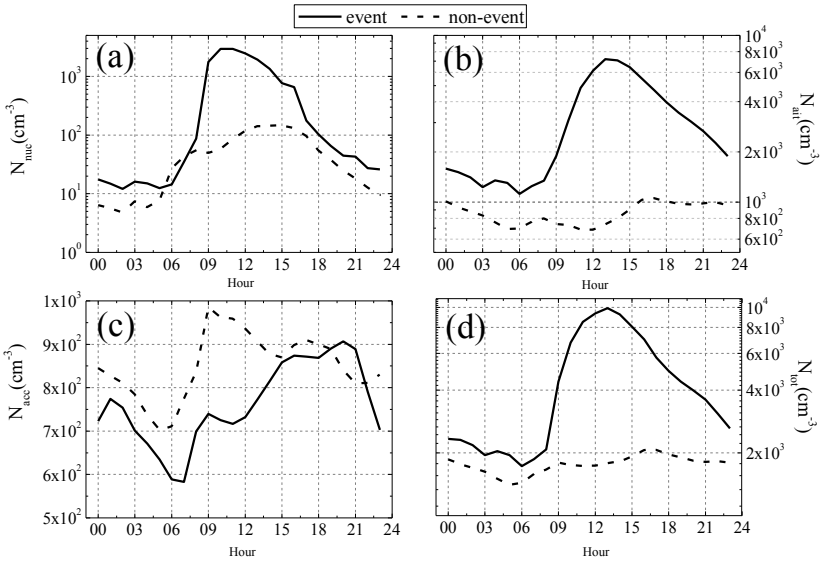

Fig 4. Comparison of diurnal variation of aerosol number concentration on event day and non-event day, Solid lines represent the event day, dotted lines represent the non-event day. (a) Nucleation mode aerosol number concentration(N_{nuc}); (b) Aitken mode aerosol number concentration(N_{ait}); (c) Accumulation mode aerosol number concentration(N_{acc}); (d) 10nm~10μm aerosol number concentration(N_{tot})

3.2.3 Statistical characteristic of meteorological factors and trace gas

New particle formation occurs on certain meteorological conditions [11], Fig5 illustrates the comparison of meteorology elements and trace gases on event days and non-event days. The diurnal variation of temperature (T) and relative humidity (RH) showed the same trend on both event days and non-event days, but T and RH were both relatively lower on event days. The average T on event days was 3.27℃ with a maximum of 4.3℃ and a minimum of 0.74℃,which both are less than non-event days respectively. The average RH on event days was 14.83% less than non-event days. New particle formation occurs on days whose temperature and RH differences between day and night were quite high, this is with the same conclusion of Mäkelä et al [29]. No matter on event days or non-event days the average wind speed showed a low value of about less than 0.5 m s^{-1}, but the wind speed on event days was slightly strong with different trend during 05:00~07:00 period.. Guo et al [24] illustrated that wind speed was high during NPF events at the mountain area in Hong Kong. The result of Zhang et al [33] shows that strong northerly winds in the urban areas of Beijing were often observed before NPF events: this is significant because winds from this direction can remove the aged aerosol in the atmosphere as there are relatively few pollutant sources on that side

of the city, which contributes to the growth of new particles after nucleation.

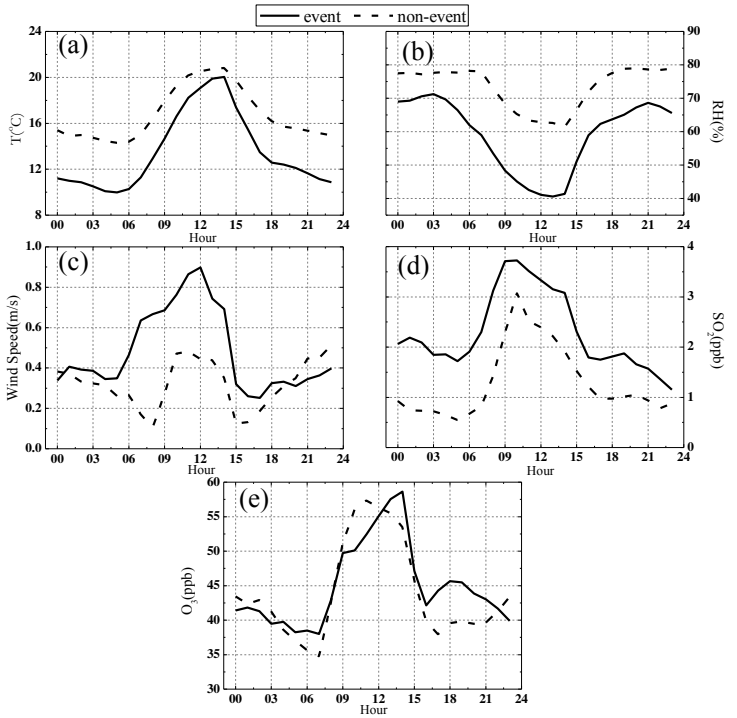

Fig 5. Comparison of diurnal variation of hourly-average meteorology elements and trace gases on event day and non-event day, (a) temperature (T); (b) Relative humidity (RH); (c) Wind Speed; (d) SO_2 concentration; (e) O_3 concentration

SO2 gas concentration at every moment on event days was higher than that on non-event days, which is same with other studies [11]. A threshold value of SO_2 is the prerequisite for the occurrence of NPF. The concentration of SO2 gas and N_{acc} on both conditions showed the same trend during 01:00~09:00 period which can be conjectured that SO2 and accumulation mode aerosol had the same source. On event days SO_2 gas concentration reached its maximum at 09:00 and maintained for 1 hour. By contrast, the SO_2 gas concentration reached its maximum at 10:00, which means the consumption of SO_2 gas during 09:00 and 10:00 on event days is larger than the increment. The consumption of SO_2 gas is due to the nucleation process of new particle formation, as is mentioned in 2.2.1, SO_2 gas may participate in the chemical reaction oxidating to H_2SO_4 and acting as the precursor. The average O_3 gas concentration was 44.92ppb on event days and 43.97ppb on non-event days. And the increment of O_3 gas concentration was also very tiny during 09:00~10:00.

3.3. The factors impact on GR

Different with each other, the averaged GR are all in a low value. As a key factor of the NPF events, GR is affected by various conditions.

Table 2 shows the statistics of the hourly average of meteorology elements, trace gases, aerosol concentrations and growth rate (GR) on event days in detail. Statistics show that the meteorology elements of temperature, relative humidity and wind speed had no direct impact on GR, but provided suitable conditions for NPF event occurrence. Fig 6 shows the positive correlation of $GR_{10\sim20}$, $GR_{20\sim30}$ and $GR_{30\sim40}$ with SO_2 concentration, and r^2 is 0.75, illustrating that SO_2 concentration is the most important factor affecting GR. The concentration of O_3 is in the range of 37~55ppb and it is 10 times higher than that of SO_2. According to eq. (2), the consumption of O_3 is half of SO_2, therefore O_3 is sufficient for new particle growth. The higher SO_2 concentration is, the more gas-phase H_2SO_4 is produced through chemical reaction. In this study, the analysis proved that H_2SO_4 is the precursor of new particle formation. Mäkelä et al [29] illustrated the source of newly formed aerosol may relate to organic activities, but it

remains to be certified through observational facts.

Table 2. Hourly average of meteorology elementes, trace gase, aerosol concentration and growth rate(GR) on event days

date	T (℃)	RH (%)	WS (ms^{-1})	SO_2 (ppb)	O_3 (ppb)	N_{nuc} (cm^{-3})	N_{ait} (cm^{-3})	N_{acc} (cm^{-3})	N_{tot} (cm^{-3})	GR (nm/h)
2012/09/30	13.88	55.13	0.44	4.11	49.05	1224.05	6122.99	1390.41	8737.45	4.27
2012/10/11	15.49	53.14	0.56	2.95	55.85	387.99	2733.27	743.77	3865.03	4.20
2012/10/12	16.29	58.42	0.32	1.94	47.42	440.58	1665.17	638.02	2743.77	3.40
2012/10/17	10.84	75.63	0.61	2.20	37.40	680.54	4071.45	771.19	5523.18	4.06
2012/10/18	11.04	62.53	0.48	1.14	41.87	515.96	2371.53	668.96	3556.45	2.29
2012/10/23	11.11	59.90	0.46	1.32	37.42	923.44	3387.75	599.13	4910.32	3.25

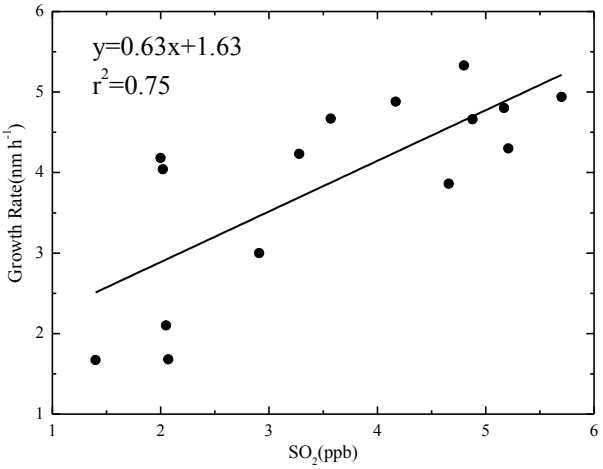

Fig 6. Correlation between particle growth rate and SO_2 concentration

4. Summary and conclusions

1. There are 6 NPF events in 33 days, accounted for 18% in total. NPF events start when the solar radiation is strong, lasting for 7~10 hours. GR is in the range of 2.29nm h^{-1} ~ 4.27nm h^{-1}. GMD and time have a good linear relationship in each NPF events, the correlation coefficients are all over 0.94.
2. The increment of aerosol number concentration shows 3 features: the time of aerosol number concentration started to increase delays with the increase of aerosol diameter; the maximum of aerosol number concentration decreases with the increase of aerosol diameter; the rate of aerosol number concentration increment decreases with the increase of aerosol diameter. Aerosol number concentration in the range of 10nm~10μm is $2.3\times10^3 cm^{-3}$ on average. The wind speed, SO_2 and O_3 concentrations are on average higher in NPF days, whereas temperature and relative humidity are at comparatively lower value.
3. SO_2 concentration has positive correlation with GR, correlation coefficient is 0.75.

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